

# **ACE-Asia Shipboard Observations of Aerosol Chemical, Physical and Optical Properties**

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## **Abstract:**

The International Global Atmospheric Chemistry Program (IGAC) has planned a series of Aerosol Characterization Experiments (ACE) that integrate in-situ measurements, satellite observations, and models to reduce the uncertainty in calculations of the climate forcing due to aerosol particles. ACE-Asia is the fourth in this series of experiments and has been designed to determine and understand the properties and controlling factors of the aerosol in the anthropogenically modified atmosphere of Eastern Asia and the Northwest Pacific. A shipboard measurement program of aerosol properties and processes is a key component in the first ACE-Asia intensive field study (<http://saga.pmel.noaa.gov/aceasia/>). This proposal seeks funding to lead the US ship component of ACE-Asia, conduct aerosol physical, chemical, and optical measurements aboard the ship during the experiment, perform aerosol chemical analyses after the experiment, and to reduce and analyze the data. The data will be made available to the research community through the NOAA Server search path at <http://saga.pmel.noaa.gov/data/> and in the ACE-Asia data archive. The data will be summarized in manuscripts that will be submitted to the JGR special sections planned for the experiment.

## PROPOSED PROJECT DESCRIPTION:

Tropospheric aerosols can have a significant impact on the Earth's radiation budget on regional scales. This is a result of aerosol properties that vary on regional rather than global scales due to relatively short aerosol lifetimes (hours to days) (IPCC(1996)). Despite the regional variability of aerosol properties, many recent estimates (e.g., IPCC(1996)) of climate forcing by aerosols rely for the most part on estimated global mean aerosol properties. Clearly, a knowledge of regional aerosol properties is needed to improve estimates of aerosol radiative forcing. This is the case for both satellite retrievals and model calculations. Inherent in satellite retrievals is the use of an aerosol model that assumes certain aerosol physical, chemical, and optical properties (e.g., Mishchenko et al., 1999). The accuracy of the retrieved aerosol properties depends on the accuracy of the assumed aerosol model. The use of time-dependent regional models built upon a knowledge of regional aerosol properties is expected to improve the accuracy of satellite retrievals (Mishchenko et al., 1999). Chemical transport models produce global aerosol distributions. Initiation and validation of the models with regionally measured aerosol properties is needed to ensure model accuracy. Radiative transfer models require values for aerosol optical properties such as the light scattering efficiency per unit aerosol mass ( $\alpha_{sp}$ ), the upward scattered fraction ( $\bar{\beta}$ ) or asymmetry factor ( $g$ ), the fraction of light scattered versus that absorbed or single scattering albedo ( $\omega_0$ ) and the dependence of scattering by the aerosol on relative humidity ( $f_{sp}(RH)$ ). All these properties depend on the chemical composition, size distribution, morphology and state of the mixture of the aerosol. Regional measurements of aerosol chemical and physical properties, including mass distributions of the dominant chemical species, the degree of mixing of various chemical species, and the number size distribution, are thus needed to link global aerosol distributions with aerosol optical properties.

Measurements across a wide range of natural and anthropogenically perturbed environments are needed to develop an accurate global picture of aerosol properties. One important region is Eastern Asia and the Northwest Pacific. Asian aerosol sources are unlike those in Europe and North America. Much more coal and biomass are burned (often with minimal emission controls), adding more absorbing soot and organic aerosol to the atmosphere (Chameides et al., 1999). Economic expansion in many Asian countries will unavoidably be accompanied by increases in fossil fuel burning. Without extensive pollution-control measures, this will increase the amount of  $SO_2$ , organic matter, and soot emitted. Furthermore, the oxidizing capacity of the East Asian atmosphere is likely to change as the growing transportation sector raises levels of nitrogen oxides (Elliott et al., 1997; van Aardenne et al., 1999). The presence of East Asia desert dust adds complexity to the regional aerosol, since it can both scatter sunlight back to space (leading to a cooling effect) and absorb solar and infrared radiation (leading to a warming effect). Mineral dust originating in Asia should be less absorbing at UV and visible wavelengths relative to the Saharan dust because of lower concentrations of iron oxides (Sokolik and Toon, 1999). However, as the mineral dust aerosol is advected eastward over the populated regions of Eastern Asia, it can become internally mixed with soot, sulfates, nitrates and/or aqueous solutions (Carmichael et al., 1996; Parungo et al., 1997). This mixed aerosol will have drastically different properties than the mineral dust near its source (Levin et al., 1996). The East Asia – Northwest Pacific aerosol is likely a complex mixture of mineral dust, carbonaceous material (black carbon and organic species), sulfates and other ionic species from urban/industrial emissions, and finally as it advects out over the ocean, sea salt. Given the

different sources, the physical, chemical and optical properties of the aerosol in this region will likely be quite different from those measured in previous ACE/INDOEX campaigns.

The radiative impact of the East Asia aerosol appears to be widespread. Based on analysis of data from the Pacific Exploratory Missions (PEM) and ACE 1 over the period 1991-1996, Thornton et al. (1999) concluded that anthropogenic sources in Eastern Asia now dominate the sulfur chemistry in the lower troposphere of the Western North Pacific eastward from the Asian continent for more than 1500 km and substantially further in the mid and upper troposphere. In addition to sulfur, the mineral dust transported from Asia can dominate the aerosol optical depth over the North Pacific (Husar et al., 1997). In situ observations on the west coast of North America confirm that transport of significant amounts of mineral dust and pollutants does occur across the expanse of the Pacific Ocean basin (Jaffe et al., 1999). The fact that much of the Asian aerosol advects out over the Pacific implies that significant changes in radiative forcing may be expected over large areas.

We propose to measure the physical, chemical and optical properties of this Asian aerosol as part of the Asian Pacific Regional Aerosol Characterization Experiment (ACE-Asia). ACE-Asia is the fourth of a series of experiments that will quantify the combined chemical and physical processes controlling the evolution and properties of the atmospheric aerosol relevant to radiative forcing and climate (<http://saga.pmel.noaa.gov/aceasia/>). The objectives of these experiments are to provide the necessary data to incorporate aerosols into global climate models and to reduce the overall uncertainty in the calculation of climate forcing by aerosols. The goal of ACE-Asia is to document the chemical, physical, and optical characteristics and determine the controlling processes of the aerosol over the polluted Asian Pacific region. This is an area strongly impacted by anthropogenic aerosols that originate in the rapidly developing Eastern Asian countries. The extent of this aerosol has been documented across the Pacific to North America (Jaffe et al., 1999).

The first ACE-Asia intensive experiment will be conducted from 15 March to 30 April 2001 in the area east of China and Japan. The project will include shipboard measurements (this proposal), aircraft measurements (NCAR-130, CIRPAS-Twin Otter, Australian and possibly Asian aircraft) and ground station measurements in China, Korea, Taiwan and Japan. The project is being coordinated by the International Global Atmospheric Chemistry Project (IGAC).

## **PROPOSAL OBJECTIVE:**

Quantify the physical, chemical, and optical properties of the atmospheric aerosol downwind of Asia under different synoptic meteorological patterns. We hypothesize that the aerosol properties downwind of ASIA will be significantly different from those that we have observed in previous field campaigns (ACE-1, ACE-2, Aerosols99, INDOEX) and that the aerosol properties will vary as a function of the prevailing synoptic meteorology and distance from shore.

## RESEARCH PLAN:

Shipboard measurements during the 2001 ACE-Asia intensive field operations ([http://saga.pmel.noaa.gov/aceasia/us\\_ship6.html](http://saga.pmel.noaa.gov/aceasia/us_ship6.html)) will contribute to the regional characterization of aerosol properties by providing data downwind of the continent. Ship operations will be directed to sample regional aerosol features (e.g. dust outbreaks, urban and industrial plumes) under different synoptic meteorological patterns and at various distances from shore. The goals of the shipboard studies are to:

- determine the physical, chemical and optical properties of the aerosol in the ACE-Asia region and assess the regional and temporal (diurnal to multi-day) variability of these properties,
- assess the major processes controlling the oxidation mechanisms of aerosol precursor gases and the formation, evolution and deposition of aerosol particles, and
- quantify the effect of the combined natural and anthropogenic aerosol on the region's radiation budget.

Our efforts, through this proposal, will focus on the first of these goals.

The cruise will be composed of two parts: an 18 day transit between Hawaii and Japan (March 10-27, 2001) and a 30 day study in the region east of Asia extending from Taiwan to Northern Japan (April, 2001). The transit will be used to quantify the gradient of aerosol properties extending from Asia to the mid-Pacific Ocean. The cruise track during the ACE-Asia intensive will depend on the synoptic meteorological conditions, the locations of the major oceanographic frontal zones, and the status of the other observational platforms and therefore must remain flexible. Several of the planned activities listed above can be carried out simultaneously as was done during ACE 1, ACE 2 and INDOEX. The ship will provide surface support for column closure experiments and perform seawater optical studies whenever meteorological conditions and aircraft/satellite schedules permit. Local closure experiments of the chemical, physical and optical aerosol properties will be conducted continuously.

### ***Proposed Measurements Funded Through this Proposal:***

#### Atmospheric Chemical Measurements

Mass size distributions of nss sulfate, sea-salt, MSA, ammonium, and other major inorganic ions with a seven stage multi-jet cascade impactor at 55% RH. IC analysis. Sampling time periods of 12 or 24 hours.

Sub- and super-micron nss sulfate, sea-salt, MSA, ammonium, and other major inorganic ions with a two stage multi-jet cascade impactor. 50% aerodynamic cut-off diameters of 1.0 and 10.0  $\mu\text{m}$  diameter at 55% RH. IC analysis. Sampling time periods of 4 to 6 hours.

Mass size distributions of elemental and organic carbon with a seven stage multi-jet cascade impactor at 55% RH. Analysis by thermal/optical transmission method (Sunset Laboratory instrument). Sampling time periods of 12 or 24 hours.

Sub- and super-micron elemental and organic carbon with a two stage multi-jet cascade impactor. 50% aerodynamic cut-off diameters of 1.0 and 10.0  $\mu\text{m}$  diameter at 55% RH. Analysis by thermal/optical transmission method. Sampling time periods of 4 to 6 hours.

Sub-micron and total elemental composition with a two stage multi-jet cascade impactor (50% aerodynamic cut-off diameter of 1.0  $\mu\text{m}$  diameter at 55% RH) and a bulk filter, respectively. XRF and SEM/XRF analysis. Sampling time periods of 4 to 6 hours.

#### Aerosol Physical and Optical Measurements

Gravimetric analysis of mass as a function of size with a seven stage multi-jet cascade impactor at 55% RH. Sampling time periods of 24 or 48 hours.

Gravimetric analysis of mass as a function of size with a two stage multi-jet cascade impactor at 55% RH. Sampling time periods of 12 hours.

Total number concentration of CN with  $D_p > 15$  nm and CN with  $D_p > 5$  nm using TSI 3010 and 3025 particle counters, respectively. Data recorded every 1 minute.

Particle number size distribution from 5 to 5000 nm diameter using an UDMPS, DMPS, and TSI 3300 APS operated at 55%RH. Data recorded every 10 minutes. (In collaboration with Dave Covert, UW)

Sub-micron and total aerosol light scattering and the backscattered fraction at wavelengths of 450, 550, and 700 nm.

Sub-micron and total aerosol light absorption at 550 nm using a PSAP.

Aerosol optical depth with hand-held MicroTops sunphotometers at 380, 440, 500, 675 and 870 nm.

#### Ancillary Measurements

Atmospheric gas phase measurements of DMS and  $\text{O}_3$

Surface air temperature, dew point, wind speed, wind direction, precipitation amount and frequency, solar insolation

Surface seawater temperature, salinity, DMS concentrations

#### ***Sampling and Analysis Methods:***

**Inlet:** Aerosol sampling and analysis methods will be similar to those used in ACE-1 (Bates et al., 1998a, 1998b; Quinn et al., 1998), ACE-2 (Bates et al., 2000; Quinn et al., 2000), Aerosols99, and INDOEX. Aerosol particles are sampled through a heated mast that extends 5 m above the aerosol measurement container. The mast is capped with a cone-shaped inlet nozzle that is rotated into the relative wind to maintain nominally isokinetic flow and minimize the loss of supermicron particles. Air is drawn through the 5 cm diameter inlet nozzle at  $1 \text{ m}^3 \text{ min}^{-1}$  and down the 20 cm diameter mast. The lower 1.5 m of the mast are heated to dry the aerosol to a relative humidity (RH) of  $55 \pm 5\%$ . This allows for constant instrumental size cuts through variations in ambient RH. Fifteen 1.9 cm diameter electrically conductive polyethylene or stainless-steel tubes extend into this heated zone to direct the air stream at flows of  $30 \text{ l min}^{-1}$  to the various aerosol sizing/counting instruments and impactors. Comparisons of the total particle count ( $D_p > 3$  nm) during intercomparisons with the NCAR C-130 and ACE-1 ground stations agreed to within 20% suggesting minimal loss of particle number in the inlet system (Weber et al., 1999). A similar comparison with the NCAR C-130 during INDOEX showed agreement to within 5%. Assessing the inlet efficiency for larger particles is more difficult. Comparisons of marine boundary layer particle extinction based on in situ measurements and total column measurements (NASA Ames suntracking sunphotometer) during ACE-2 agreed to within the uncertainties of the measurements and calculations, however those uncertainties are quite high. Our plan before ACE-Asia, is to characterize the passing efficiency of the inlet for particles in

the 0.8 to 10  $\mu\text{m}$  diameter size range. This project will be conducted in the University of Washington's Kirsten Wind Tunnel with funding from a separate proposal.

**Number concentration and size distribution data:** Data will be filtered to eliminate periods of calibration and instrument malfunction and periods of ship contamination (based on relative wind and high and rapid changes in CN counts). The filtered mobility distributions from the DMPSs will be converted to number-size distributions using the inversion routine of Stratman and Wiedensohler (1997). The data will be corrected for diffusional losses (Covert et al., 1997) and size dependent counting efficiencies (Wiedensohler et al., 1997) based on pre-ACE-1 and ACE-2 intercalibration exercises. The number size distribution between 0.6 and 9.6  $\mu\text{m}$  will be measured with an aerodynamic particle sizer (APS) at an RH of 55%. The APS diameters will be converted to geometric diameters by dividing by the square root of the particle density determined from the size-resolved chemical measurements. The number size distribution will be measured at 55% RH to be directly comparable with the impactor size cuts and optical measurements. For comparison with previous data, the number size distribution will be adjusted to 10% RH based on the growth factors measured on the ship (separate proposal from David Covert). An interactive routine will be used to fit lognormal curves to the different modes of the number size distribution (Bates et al., 2000). The sizing instruments will be tested against the ACE-Asia sizing calibration standard at the CIRPAS in Monterey California prior to the experiment.

**Mass size distributions:** Seven-stage multi-jet cascade impactors (Berner et al., 1979) sampling air at 55% RH will be used to determine the mass size distributions of  $\text{Cl}^-$ ,  $\text{Br}^-$ ,  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$ , methanesulfonate ( $\text{MSA}^-$ ),  $\text{Na}^+$ ,  $\text{NH}_4^+$ ,  $\text{K}^+$ ,  $\text{Mg}^{+2}$ , and  $\text{Ca}^{+2}$ . Sampling at 55% RH has been shown to eliminate particle bounce for ammonium sulfate salts and sea salt (Quinn, unpublished data). The 50% aerodynamic cutoff diameters,  $D_{50,\text{aero}}$ , are 0.18, 0.31, 0.55, 1.1, 2.0, 4.1, and 10  $\mu\text{m}$  at 30 lpm. The impaction stage at the inlet of the impactor will be covered with silicone grease to prevent the bounce of large particles onto the downstream stages. Tedlar films will be used on the largest stages of the impactor and a Millipore Fluoropore filter (1.0- $\mu\text{m}$  pore size) will be used for the smallest stage. All handling of the substrates will be done in an  $\text{NH}_3$ - and  $\text{SO}_2$ -free glove box. Blank levels will be determined by loading an impactor with substrates but not drawing any air through it. Similar measurements will be made on samples collected with two-stage multi-jet cascade impactors sampling air at 55% RH to provide shorter time-resolution samples. The impactor has  $D_{50,\text{aero}}$  of 1.1 and 10  $\mu\text{m}$  at 30 lpm. NOAA/PMEL will lead the ACE-Asia inorganic ion standard intercomparison during the summer of 2000.

Seven and two-stage multi-jet cascade impactors sampling air at 55% RH will be used to determine concentrations of total, organic, and elemental carbon. Particles will be collected on Al foils and a quartz filter. Positive artifacts associated with collecting gas phase organic species will be assessed using a backup quartz filter placed behind the quartz filter in this impactor and the Fluoropore filter in the impactor used for ionic measurements. The Al foils and quartz filters will be combusted immediately before use at 600°C for 4 hours to remove organic contaminants. Blank levels will be determined by placing substrates into a second impactor and deploying the impactor for the duration of the sampling period without drawing air through it. Foils and filters will be stored frozen in capped glass vials until analysis. The organic analysis will be done using a thermal-optical technique (Sunset laboratory instrument, Birch and Cary, 1996). The method uses a constant monitoring of the optical absorbance of the sample during analysis to correct for the carbon-char formed by the pyrolytic conversion of some organic species into additional elemental carbon. As with all current techniques for organic/elemental carbon, this is a method-

defined analysis. However, the data should be directly comparable with the shipboard carbon measurements made in ACE-2 (Novakov et al., 2000) and Aerosols99/INDOEX (samples currently being run at IFT in Leipzig, Germany). We will plan to participate in the pre-campaign intercomparison tentatively scheduled for late summer of 2000.

**Elemental analysis and particle morphology and mixing state:** Total elemental composition (Na, Mg, Al, Si, P, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, Ba, As, and Pb) will be determined by thin-film x-ray primary- and secondary-emission spectrometry. Our laboratory is currently equipped with a Kevex Model 8000-770 x-ray energy spectrometer. However, prior to ACE Asia this instrument will be replaced with either an EX-6600AF system manufactured by Jordon Valley or a Spectro X-Lab 2000 from Asoma Instruments, Inc.

Particle mixing state and morphology will be determined using an ISI DS130S Scanning Electron Microscope equipped with a Kevex SuperQuantum Extra detector with an ultra thin window used at 6 – 20 kV and from 300 to 65000X (Feely et al., 1998). Semi quantitative analysis will be performed with an IXRF Systems Model 500 Analyzer. Particle size analysis of individual particles and aggregates in the fields will be determined with IXRF's Iridium software. For both the XRF and SEM analysis, submicron samples will be collected on the after filter of a two-stage multi-jet cascade impactor. Total samples will be collected with a bulk filter. This method allows for a sharp size cut between the sub- and supermicron particles for the XRF analysis and different sampling times for the two size ranges as often required by the SEM analysis.

**Gravimetric analysis:** Two and seven-stage multi-jet cascade impactors sampling air at 55% RH will be used to collect aerosols for gravimetric analysis (Quinn and Coffman, 1998). A 47-mm Millipore Fluoropore filter will be used for the after filter and 90-mm Fluoropore films will be used for the impaction stages. Filters and films will be weighed at PMEL before and after sample collection with a Mettler UMT2 and Cahn Model 29 microbalance, respectively. The microbalances will be housed in a glove box kept at a humidity of 55±2%.

**Scattering and absorption:** Measurements of aerosol scattering and backscattering coefficients will be made with an integrating nephelometer (Model 3563, TSI Inc.) at wavelengths of 450, 550, and 700 nm. Data will be corrected for angular truncation errors using the method of Anderson and Ogren (1998). The absorption coefficient will be measured at 550 nm by monitoring the change in transmission through a filter with a Particle Soot Absorption Photometer (PSAP, Radiance Research). Data will be corrected for scattering artifacts, spot size, and manufacturer's calibration using the method of Bond et al. (1999). Both the nephelometer and the PSAP data will be reported at STP. Two single-stage cascade impactors, one having a  $D_{50,aero}$  of 1.1  $\mu\text{m}$  and the other 10  $\mu\text{m}$ , will be placed upstream of the nephelometer and PSAP. A valve will be switched between the two impactors every 15 min so that sampling alternates between submicron aerosol and sub-10  $\mu\text{m}$  aerosol. Scattering, backscattering, and absorption by the supermicron aerosol will be determined by difference. The scattering, backscattering, and absorption coefficients will be measured at 55% RH.

### ***Post-cruise Sample and Data Analysis:***

1. Complete the chemical and gravimetric data analysis and data reduction. Most of the ion chromatography analysis will be done onboard ship however the samples that are returned to the laboratory for gravimetric analysis will also be analyzed by ion chromatography. Samples will be run for organic/elemental carbon, XRF and XRF/SEM.

2. Complete the data reduction of the physical size distribution and optical measurements. The measured chemistry will be used in the PMEL chemical equilibrium model to calculate aerosol densities and refractive indices. The densities will be used to shift the APS data to geometric diameters. The APS data will then be combined with the DMPS data and lognormal curves will be fit to the several modes of the number size distribution. Output from the model also will be used to calculate scattering and backscattering coefficients for comparison to measured values and to partition scattering and optical depth amongst the measured chemical components.
3. Complete the data reduction of the ancillary data, calculate air-mass back trajectories along the cruise track, and provide these data sets to other cruise participants.

### ***Data Products:***

The objective of the proposed research is to quantify the physical, chemical, and optical properties of atmospheric aerosol downwind of Asia under different synoptic meteorological conditions. We hypothesize that the aerosol properties downwind of Asia will be significantly different than those we have observed in previous field campaigns and that the aerosol properties will vary as a function of the synoptic meteorology and distance from shore.

To achieve this objective, our data analysis will initially include the following:

1. The data sets will be categorized according to synoptic meteorological patterns (based on surface weather maps, meteorological data, and back trajectories) and distance from shore.
2. For each category, we will calculate the mean, variability and uncertainties of the:
  - a) Number and surface area modal parameters
  - b) Absolute concentrations and sub- and supermicron mass fractions of the major chemical components
  - c) Optical properties including the scattering coefficient, backscattered fraction, Angstr $\star$ m exponent, absorption coefficient, single scattering albedo, aerosol optical depth
3. SEM/XRF analysis will be used to assess the degree of mixing of various chemical species and morphology.
4. Aerosol scattering and aerosol optical depth (using surface chemistry measurements and lidar information) will be apportioned to the different chemical components.
5. The factors (size and chemistry) that control aerosol optical properties in the different synoptic patterns and with distance from shore will be assessed.

In addition, we will:

6. Provide the data to the research community through the NOAA Server search path at <http://saga.pmel.noaa.gov/data/> and the ACE-Asia data archive. Maintain the project web page.
7. Participate in ACE-Asia data workshops to intercompare the data sets from the various platforms and to integrate the overall data set.
8. Participate in the ACE-Asia special sessions at AGU and the CACGP/IGAC meetings.

9. Prepare manuscripts for the ACE-Asia special sections of JGR-Atmospheres. Based on our experience with ACE 1 and ACE 2 (see results from prior research), we anticipate that our data sets also will be widely used by other PIs in their data analyses.

Examples of data products resulting from the approach outlined above are shown in Figures 1 and 2. Frequency distributions of single scattering albedo for different regions are shown in Figure 1. Single scattering albedo is a measure of the absorbing versus the scattering nature of the aerosol. The lower the value, the more absorbing the aerosol. ACE 1 values are clustered near one confirming a composition of sea salt and sulfate with little black carbon. The distribution of values for ACE 2 and TARFOX is broader and extends to much lower values indicating a mix of marine aerosol composed of scattering species and continental aerosol composed, in part, of black carbon. The INDOEX frequency distribution is interesting in that it peaks at lower values than either ACE 2 or TARFOX. This is a result of black carbon-laden aerosol advecting off of the Indian subcontinent. Average number size distributions sorted by air mass trajectory during ACE 1, ACE 2, Aerosols99 and INDOEX are shown in Figure 2. The modal parameters are clearly different as a result of the source, transport and subsequent aerosol transformation. These analyses clearly show regional differences in aerosol properties that strongly influence the radiative forcing by the aerosol.

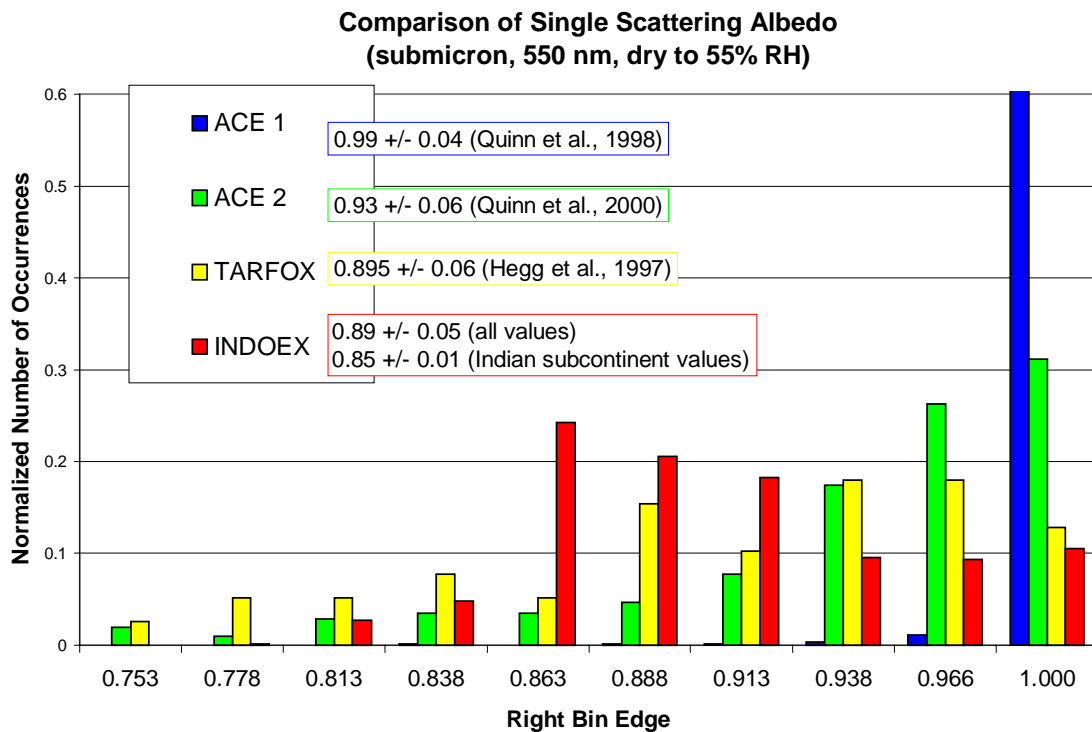


Figure 1. Frequency distributions of single scattering albedo measured during 4 field experiments.

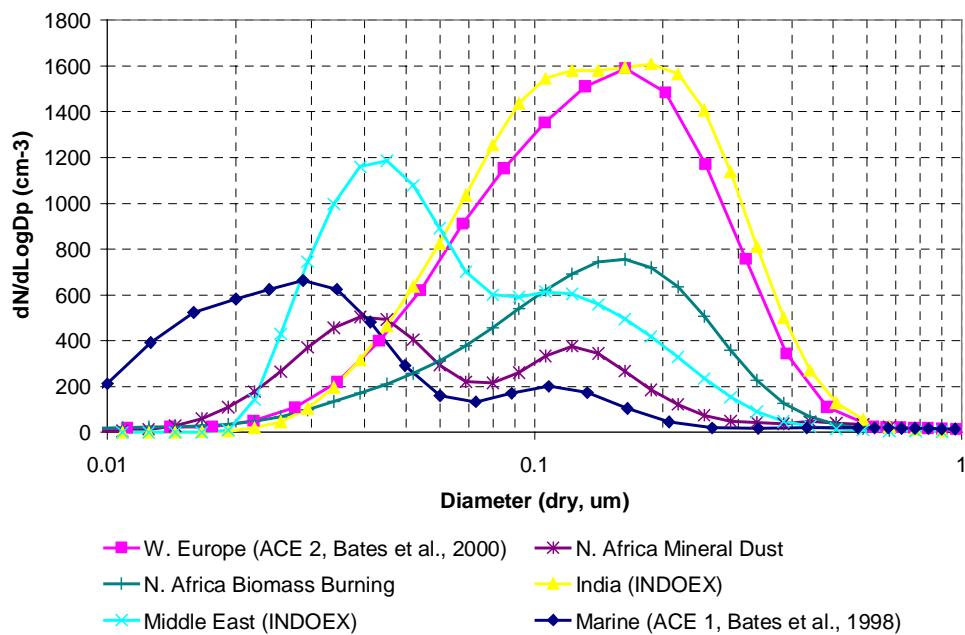


Figure 2. Average number size distributions sorted by air mass trajectory for ACE-1, ACE-2, Aerosols99 and INDOEX

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